

Power thresholds of morphology dependent induced thermal scattering in silica microresonators

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Abstract

Induced thermal scattering power thresholds have been calculated as a function of size and laser pump frequency. The thermal coupling coefficients of morphology dependent resonances were estimated by asymptotic methods. The resulting power threshold is comparable with experimental observations of thresholds of Raman lasing and thermal instability in spherical silica resonators. Applications may include the remote measurement of the temperature of aerosol droplets and the stabilization of microcavity lasers against thermal oscillations and temperature deviations on microcavity. A silica resonator can be used as an IR sensor, as well as an additional tool for precisely measuring the thermal conductivity and heat capacity of a target in a microsphere by calculating of the thermal shifts of eigenfrequencies in spectra of nonlinear scattering.

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Numerous stimulated nonlinear optical processes, including Mandelstam-Brillouin and Raman scattering have been observed at very low threshold laser pump intensities in liquid and solid microresonators[1, 4]. The main explanation for the lowering of threshold conditions in microresonators is presented in Ref.[5, 6]. The nonlinear solid state silica microresonators, having the "Whispering Gallery Modes" (WGM) with high Q-factor ($Q = 10^8$), act as low threshold microsphere Raman lasers[2, 3]. The life time of photons of WGM is significantly longer than in bulk solid. Due to the high photon number in the effective volume of WGM and the long life time of photons at "Morphology Dependent Resonance" (MDR) conditions, there is a low Raman lasing threshold, which was observed in the experiments with silica spheres having radius $R = 40 \mu m$ [2]. The threshold power of the laser pump is $86 \mu W$ for a pump with wavelength $1.55 \mu m$, which is quite low[2]. The absorbed pump power at MDR conditions changes the eigenfrequency of a microresonator by heating the effective volume of WGM. A periodic shift of the eigenfrequency provides thermal instability in microresonators[10] and leads to a bistability in the cavity[8, 9]. At the specific threshold intensity of the laser pump and tuning conditions, the resonator surface layer can significantly enhance the internal fields at MDR's and efficiently provide optical thermal feedback for the internally generated WGM, which leads to optical bistability and instability effects[7, 8, 9, 10]. The low threshold power of thermal bistability and thermal oscillation in silica microresonator have been reported for silica resonators with radii $100 \mu m$ and $150 \mu m$ at pump wavelength $0.63 \mu m$ [10]. The high Q-factor of WGM provides the lowering threshold conditions for the laser induced thermal scattering in the microresonator. Laser stimulated thermal scattering in liquid bulk have been reported in Ref.[11, 12]. The goal of this article is to provide a theoretical calculation of the threshold power of the laser induced thermal scattering (LITS) in support of recent experimental efforts in the area of thermal nonlinear effects in microresonators[8, 9]. LITS is described by a system of equations for nonlinear oscillators with partial wave electromagnetic amplitudes TE_n^1 and thermal modes $T_n^{0,1}$ in the resonator[13]. There are numerous regimes of LITS depending on the combinations of the mode overlapping and the amplitudes of partial electromagnetic and thermal modes; these regimes include thermal instability, thermal self-modulation and aperiodic thermal oscillation or bistability[7, 8, 9]. Experimentally, the effect of thermal instability takes place in ring resonators[8, 9] or fused silica spherical resonators[10]. There are two interacting modes within the homogeneous thermal mode volume, one being the pump

mode and one being the resonant signal (anti-Stokes) mode, i.e. the input and the output resonance conditions (MDR's). The input resonance condition is satisfied for a broadband thermal detuning η of the pump input signal, which spans several high-Q MDR's, whereas the output resonance condition is always satisfied, since the bandwidth of LITS spans at least several high-Q MDR's. The high-Q factor modes are modulated by the temperature oscillations of the resonator due to the thermal shift of MDR's peaks near ω_f . The relevant Lorentzian is modulated by Ω_T and by the amplitudes corresponding to the thermal oscillations. In the case of the interaction of two modes (one a thermal mode T_1^1 and the second mode a WGM mode TE_n^1), the threshold power is taken into account by applying the methods of slowly varied amplitudes for the system of ordinary differential equations describing the oscillations. The power threshold takes the form [13]:

$$P_{th} = \frac{\omega_p \rho C_p V}{2a_\varepsilon Q_f^2} \frac{1 + \tau}{1 \pm \gamma} \frac{1 + \eta^2}{2\eta} \quad (1)$$

where:

$$\gamma = \frac{3\pi \chi^{(3)} \varepsilon^2 \rho C_p}{\omega_f a_\varepsilon} \quad (2)$$

is a thermal and electromagnetic mode coupling coefficient and

$$\tau = \frac{2KQ_f}{\rho C_p \omega_p} \left(\frac{\mu_j}{R} \right)^2 \quad (3)$$

The thermal anti-Stokes frequency has the form[13]:

$$\Omega_T = \omega_p \left[\frac{\xi_f^2/4 + (\tau_t + \tau_e)^2}{1 - \gamma} - \tau_t^2 \right]^{1/2} \quad (4)$$

where: ω_p is the laser pump frequency, $\eta = \xi_f/\xi_0$, $\xi_f = 1 - \omega_f^2/\omega_p^2$ and $\xi_0 = 2(\tau_t + \tau_e)$ is the optimal detuning, $\tau_t = K(\mu_i/R)^2/\rho C_p \omega_p$ and $\tau_e = 1/2Q_f$ are the dimensionless time of the thermal and the electrical relaxation of the WGM, V is the WGM's volume[10], μ_i are the roots of the boundary secular equation for the spherical geometry of the resonator[13]. In the result that follows, it is assumed that the pump field is intense and undepleted, as opposed to the anti-Stokes field. We can now derive the threshold incident power P_{th} of the pump for LITS using the basic relation for any Q-factor, namely, that a Q-factor is the ratio of the field energy inside the mode to the incident power, multiplied by the leakage rate. The best condition for observation of LITS is provided by a spherical resonator from fused

quartz with a high Q-factor for WGM. The threshold incident power P_{th} of the pump for LITS can be derived using the basic relation for any Q-factor namely,

$$\frac{1}{Q_f} = \left(\frac{1}{Q_{scat}} + \frac{1}{Q_{abs}} \right)_f \quad (5)$$

where $(Q_{scat})_f$ and $(Q_{abs})_f$ are the Q-factors of scattering loss and absorption loss in the f-mode. To be specific, here we concentrate on the calculation of the threshold power of LITS for modes whose amplitudes and resonance half-widths and threshold of Raman lasing are given in [2, 4]. The calculated threshold power P_{th} and the anti-Stokes frequency Ω_T are presented by Fig.1-6, using the known values of material parameters for the fused silica valid for the experiments: the density of fused silica $\rho = 2.21[g/cm^3]$, the thermal conductivity $K = 1.4 \cdot 10^{-2}[W/cmK]$, the specific heat capacity $C_p = 0.67[Ws/gK]$, $a_\epsilon = 1.45 \cdot 10^{-5}[K^{-1}]$, third-order susceptibility $\chi^{(3)} = 5 \cdot 10^{-15}[esu]$, $n = \epsilon^2$, the refraction index $n = 1.46$. The threshold power of LITS is then determined in two cases:(i) effective resonant heat absorption and thermal mode effective overlapping, and (ii) heat exchange on the surface of the resonator. The next assumption takes into account the small losses of the electromagnetic mode, which corresponds to the WGM's having a high-Q factor[10, 15]. Optimal tuning conditions for observing of LITS can be created using a spherical resonator from fused silica with a high Q-factor of $Q_f = 10^7$ [15]. The Q-factor in Eq.(1) and Eq.(4) leads to the expected linear dependence of the loss rate. The absorbed pump energy depends on the laser pump power, while the Q-factor leads to LITS through the contribution of stored energy within the electromagnetic mode volume, which heats the system and raises the entropy through the contribution of the temperature dependence of the index of refraction on the scattering volume. This dependence is reminiscent of stimulated thermal scattering in liquid bulk and is a important factor in understanding the single-photon absorption rate for a molecular system with a chemical impurity. Thus, the threshold power of the incident laser pump is inversely proportional to the thermal coupling of partial modes γ , and to the Q-factor of the pump mode squared. The LITS power threshold evaluated for the optimal detuning ξ_0 in MDR's as a function of resonant pump wavelength is presented in Fig.1. The interacting electromagnetic modes E_n^1 corresponding to WGM's $n \approx \rho$, where: $\rho = 2\pi R/\lambda_p$ is the size parameter, R - radius of the resonator, λ_p is the pump wavelength. The threshold power and thermal combination anti-Stokes frequency with dependence from detuning ξ_f presented in Fig.2 and Fig.3. As illustrated in Fig.2-3, the optimal tuning con-

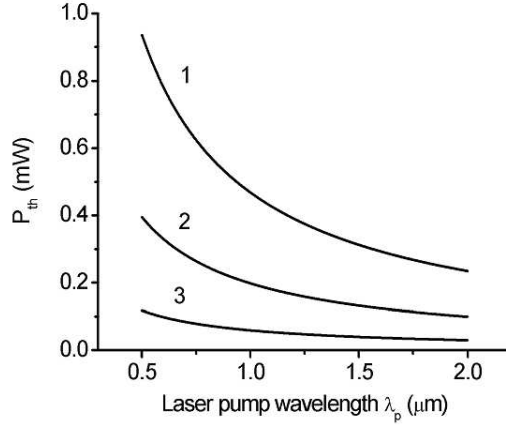


FIG. 1: The threshold power of LITS depends on resonant wavelength of wide range of gas laser and solid state semiconductor lasers. Radius of fused silica microsphere: 1. $R = 20 \mu m$, 2. $R = 15 \mu m$, 3. $R = 10 \mu m$, $Q_f = 10^8$, $TE_n^1 - T_1^1, n \approx \rho$.

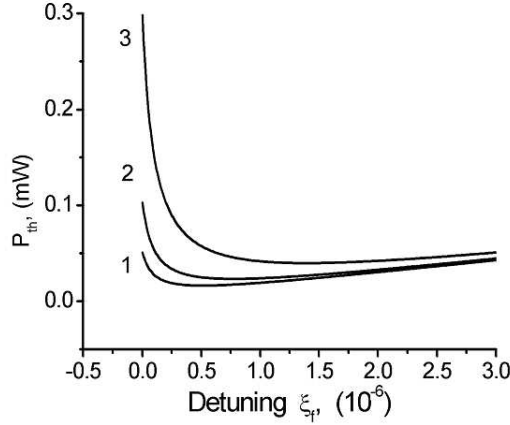


FIG. 2: The threshold power of LITS as a function of detuning ξ_f (radius $R = 35 \mu m$) exited the MDR's: 1. $\lambda_p = 0.532 \mu m$, $TE_{600}^1 - T_1^0$ modes, 2. $\lambda_p = 0.840 \mu m$, $TE_{300}^1 - T_1^0$ modes. 3. $\lambda_p = 1.55 \mu m$, $TE_{210}^1 - T_1^0$.

dition provides the minimum of incident laser power for LITS inside a microresonator. It corresponds to the detuning of the effective two-mode interaction with ξ_f comparable with the ξ_0 . The threshold power has a minimum at the optimal tuning of the pump wavelength. The threshold power of LITS, as a function that depends on the laser pump wavelength, is presented in Fig.4. It was included in the detuning function ξ_f . The minimum of the threshold intensity is achieved by MDR's tuning at $\lambda_p = 0.532 \mu m$ and consists of $20 \mu W$.

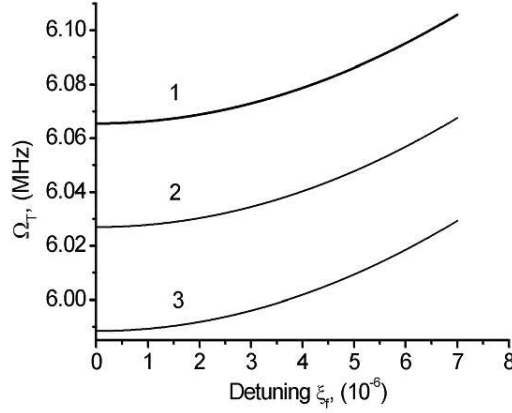


FIG. 3: The thermal combination anti-Stokes frequency of LITS as a function of detuning ξ_f (radius $R = 35 \mu m$) exited the MDR's: 1. $\lambda_p = 0.532 \mu m$, $TE_{600}^1 - T_1^0$ modes, 2. $\lambda_p = 0.840 \mu m$, $TE_{300}^1 - T_1^0$ modes. 3. $\lambda_p = 1.55 \mu m$, $TE_{210}^1 - T_1^0$.

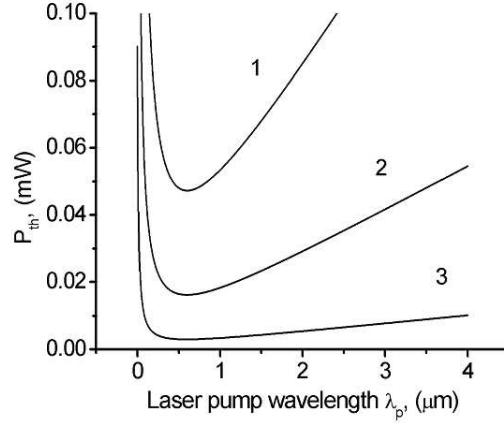


FIG. 4: The minimum of threshold of LITS for silica microsphere at the laser pump $\lambda_p = 0.532 \mu m$ with radii 1. $R = 70 \mu m$, 2. $R = 45 \mu m$, 3. $R = 50 \mu m$.

It is consistent with the experimental data[10] and less than that of the threshold for Raman lasing[2]. The computed threshold for input power in a microresonator varies from 20 to 50 μW for detunings of 5 to 15 linewidths from a TE resonant mode. Our new result for threshold power and Ω_T calculated by the formulas (1)-(4) are presented in Fig. 5 and Fig. 6 allowing us to compare the threshold and combination frequencies of relevant experimental parameters for the small spherical silica particles suspended or spraying in the atmosphere. The Ω_T for the microspheres with radii $R = 2 \mu m \div 10 \mu m$ (the probe

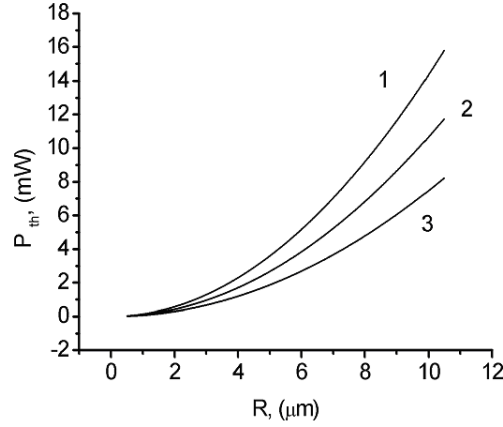


FIG. 5: The threshold power of LITS for fused silica microsphere. $Q_f = 10^7$, $\lambda_p = 1.550 \mu m$, 1. $TE_{230}^1 - T_1^0$, 2. $TE_{170}^1 - T_1^0$, 3. $TE_{120}^1 - T_1^0$.

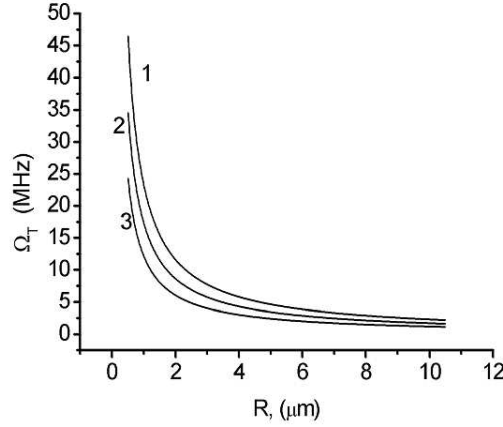


FIG. 6: The thermal combination anti-Stokes frequency of LITS for fused silica microsphere $Q_f = 10^7$, $\lambda_p = 1.550 \mu m$, 1. $TE_{230}^1 - T_1^0$, 2. $TE_{170}^1 - T_1^0$, 3. $TE_{120}^1 - T_1^0$.

particles in the atmosphere) is smaller than $5 MHz$ in the regime of time of pump pulse: $\tau_p \gg \tau_t \gg \tau_e$. The thermal combination frequency Ω_p for the silica spheres with radius of $5 \mu m$ has a value between $5 MHz$ and $10 MHz$. For a micrometer-sized silica resonator the thermal mechanisms become important for time scales on the order of τ_t for thermal relaxation, a few microseconds. It can occur in the Raman and Mandelstam-Brillouin spectra in the silica spheres and can provide thermal modulation of Raman and Mandelstam-Brillouin scattering amplitudes in the experiments with lasing in silica spheres. The combination of anti-Stokes frequency of LITS can be varied in a wide range from a value of combination

Rayleigh frequencies (Rayleigh thermal scattering[11, 12]) to striction Brillouin frequencies (Mandelshtam-Brillouin scattering[4]). Such a wide variation of values of anti-Stokes frequency Ω_T is provided by the distributed nonlinear system, like a resonator. The calculated threshold power of LITS is significantly less than the thresholds of Mandelstam-Brillouin scattering, which is less than 160 W[4]. Thus, it is necessary to take into account LITS on small particles in the scattering experiments. It is found that the thresholds of parametric LITS in silica microspheres are amenable to experimental observation, because this threshold is commensurate with the threshold of stimulated Raman scattering on microspheres or also with the threshold Raman lasing in the volume of WGM at resonator[2]. Furthermore, we may assume that the normalized gain of Raman lasing can be modulated by the excitation of thermal mode of higher order by partial electromagnetic waves. Thermal modes and the surface WGM mode pairs with strong nonlinear coupling have low thresholds and thermal combination frequencies $2.5 \div 50$ MHz. As was shown in[16] there is a line broadening of the modes if the microsphere is doped with latex nanoparticles. The concentration of any absorbing or scattering nanoparticles can be estimated by power thresholds and Anti-Stokes frequency shifts with size parameters. It provides the experimental tool for controlling the polymerization on the surface of microdroplets and microresonators. The threshold for LITS will decrease due to losses incurred by absorbing nanoparticles. However, if the nanoparticles are present on the surface of resonators, the effect of stimulated Raman scattering, Raman lasing and line-broadening may keep the thermal modulation of Q-factors intact and cause the splitting of resonant modes. In this case the integral coefficients of interaction between Anti-Stokes and pump (or signal and idler) modes may actually increase. This intriguing possibility leads to further lowering of the threshold conditions of LITS, whose practical aim may be the creation of broad-band microsphere thermal sensors. This work has provided the asymptotical an approach for threshold conditions of LITS. In order to run more exact calculations, it may be necessary to develop a detailed theory of thermal and electromagnetic mode overlapping. The theory presented above could be used for resonators of any geometrical form, but it while necessary to provide the more complicated solution for boundary problems of diffraction and thermal conductivity theory, even though the basic physics of our solution does not change. Threshold measurements can be an effective tool

for chemical substance detection and for the creation of IR sensors.

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